

# PATENT SPECIFICATION

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## DRAWINGS ATTACHED

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## (54) FUEL CELL DEVICE

(71) We, ALLMÄNNA SVENSKA ELEKTRISKA AKTIEBOLAGET, a Swedish company, of Vasteras, Sweden, do hereby declare the invention, for which we pray that a Patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following Statement:—

This invention relates to a fuel cell device. Air is a particularly attractive oxidant for a fuel cell. This is of course so for all fuel cell systems which operate in a free air *milieu*, for example traction systems such as cars, locomotives, trucks and stationary systems such as small power stations for telecommunication links, beacons, buoys and automatic weather stations. When air is used as oxidant the difficulty arises, especially for small fuel cell systems, of maintaining the pressure difference between the air side and the electrolyte side of the air electrode (which is necessary so that the pores of the porous electrode will not be completely filled with liquid and the electrode therefore lose practically all activity) without the help of considerable auxiliary power which is not conveniently available in small systems.

Usually the electrode is made hydrophobic, which may be done, for example, by coating the pores with wax or paraffin or by adding polytetrafluorethylene particles to the powder from which the porous electrode is manufactured. The electrolyte is thereby prevented from passing right through the electrode to the atmosphere and the menisci of the electrolyte assume a position of equilibrium in the porous electrode. During long operations, however, difficulties often arise since the effect of the hydrophobic substance decreases (for example due to the formation of pore coatings or chemical reactions) so that the electrode eventually becomes completely filled with the electrolyte. This "ageing" phenomenon has been found to be particularly pronounced with the use of fuel, for example methanol, dissolved in the electrolyte, since the surface tension of the electrolyte is then substantially reduced. An undesirable

effect is also obtained when hydrophobic material is used since the hydrophobic material partly screens the catalyst surface. The use of polytetrafluorethylene as a hydrophobic material also means that a lower sintering temperature must be used when the electrode is manufactured, which in turn means that the desired mechanical strength of the electrode cannot be achieved.

The required pressure difference between the two sides of the air electrode to prevent total absorption of liquid can also be maintained by supporting a capillary active membrane against the electrode on the electrolyte side. Such a membrane may consist, for example, of asbestos or some other porous material having extremely fine pores. In this way the electrolyte is prevented by "blotting paper effect" from penetrating through the electrode. Even with this arrangement the capillary active effect may decrease with long operating times for just those reasons discussed for the case of hydrophobic electrodes. A disadvantage also arises with fuel cells having circulating electrolyte in that the electrolyte resistance increases strongly so that the flow of the electrolyte through the electrolyte chamber is greatly impeded.

The present invention aims at providing a fuel cell system which is self-sufficient as far as its energy requirements are concerned for a considerable time and which can operate for long periods without unfavourable alterations taking place in the electrodes.

According to the present invention a fuel cell device comprises at least one fuel cell having a fluid electrolyte arranged in an electrolyte chamber, a fuel electrode arranged in contact with the electrolyte and with the fuel of the fuel cell and an air electrode arranged in contact with the electrolyte and with the atmosphere, the air electrode having one side contacting the atmosphere and one side contacting the electrolyte and comprising at least two porous layers having different porosity, the layer situated nearest the electrolyte having finer pores than the layer situated nearest the atmosphere, the elec-

[Price 25p]

trolyte chamber being connected by an openable and closable conduit to an evacuated container which can be maintained at a pressure lower than atmospheric pressure by means of an evacuating pump connected to the container by an openable and closable conduit, the pump being arranged to be operated intermittently by electric energy generated in the fuel cell, the electrolyte being arranged in an electrolyte circuit containing an electrolyte storage vessel having a volume which is large in relation to that of the electrolyte chamber.

The pressure difference between the atmosphere side and the electrolyte side necessary for the function of the air electrode can be maintained for long periods (for example the time between two operations for supplying fuel using fuel dissolved in the electrolyte) if the evacuated container is made very large in relation to the quantity of gas entering into the electrolyte chamber from outside.

The intermittent operation of the evacuating pump means that only a very small part of the electrical energy generated in the fuel cell is used to maintain the pressure difference. The container can be evacuated during operation of the fuel cell since it is connected to the fuel cell by an openable and closable conduit.

As the electrolyte is arranged in an electrolyte circuit containing an electrolyte storage vessel, renewal of the electrolyte in the fuel cell can be effected by circulation of the electrolyte in the circuit whenever this is required. This is important particularly for fuel cells employing fuel dissolved in the electrolyte because the reserves of electrolyte and fuel can be so large that fresh fuel does not need to be supplied until after a long period of operation.

In the layer of the air electrode which is situated on the atmosphere side, that is the layer with the larger pores; the average pore diameter is so large, preferably 10—100  $\mu$ , that a relatively low pressure difference can be used as the optimum operating pressure, for example 0.05—0.6 atm. and preferably 0.1—0.4 atm. In the layer facing the electrolyte, that is the layer with the finer pores, the diameter of the largest pores is preferably 1—30  $\mu$ . By making the latter layer with a thickness of at least 0.2 mm and a largest pore diameter which is smaller than the average pore diameter of the active layer, the risk of air bubbling through to the electrolyte is practically non-existent at reasonable pressure differences.

If a gaseous fuel, for example pure hydrogen gas or a gas mixture containing hydrogen gas and, in addition, for example nitrogen gas or carbon dioxide gas are added to the fuel electrode, this is also given two porous layers, one facing the fuel side and provided with larger pores and the other facing the

electrolyte side and provided with finer pores. For these two layers, pore diameters and thickness are chosen in accordance with what has been stated for the corresponding layers in the case of an air electrode.

If a fuel dissolved in the electrolyte is used the fuel electrode need not, of course, consist of two layers. It is then made porous to a suitable extent and consists preferably of only one layer. Suitable fuels which can be used dissolved in the preferably alkaline electrolyte are among others methanol, other low monofunctional alcohols, glycol, formic acid, a formate or hydrazine. Methanol, formic acid or a formate are particularly preferred for economic and practical reasons.

The electrolyte preferably consists of an alkali metal hydroxide such as an aqueous solution of potassium hydroxide containing 1—35 per cent by weight of the hydroxide.

The invention will now be further described, by way of example with reference to the accompanying drawing, in which:—

Figure 1 illustrates a fuel cell device according to the invention employing a fuel dissolved in the electrolyte and,

Figure 2 illustrates a fuel cell device according to the invention employing a gaseous fuel.

Referring to Figure 1 a fuel cell 1 has a fuel electrode 2 and an air electrode 3. The fuel electrode 2 consists of a porous plate of sintered nickel particles. On the electrode and in its pores, which have an average diameter of 10  $\mu$ , is a coating consisting of a mixture of palladium, platinum and ruthenium. The platinum metals may be applied in a quantity of 0.01—5 per cent, for example 0.5 per cent, of the weight of the electrode. The thickness of the plate is about 1.5 mm.

The air electrode 3 consists of an active layer 4 for the electrode reaction, having larger pores and an inactive layer 5 with smaller pores. The active layer 4 consists of a mixture of silver and nickel particles sintered together, in which the silver comprises 40 per cent by weight and the nickel 60 per cent by weight. The average diameter of the pores is about 35  $\mu$  and the thickness of the layer is about 1.5 mm. The inactive layer 5 consists of nickel particles which have been compressed and then sintered on the layer 4. The diameter of the largest pores in this layer is about 10  $\mu$  and the thickness of the layer is about 0.4 mm.

An electrolyte 7 arranged in an electrolyte chamber 6 consists of an aqueous solution of potassium hydroxide containing 20—25 per cent by weight KOH. The electrolyte also contains methanol or formic acid, which forms the fuel of the fuel cell, in a percentage of 10%, calculated on the total weight of water, potassium hydroxide and methanol. The electrolyte on both sides of the fuel

electrode 2 is in contact through the pores of the electrode.

The air electrode is in contact with the atmosphere outside the layer 4.

5 The electrolyte chamber 6 is in contact through conduits 8 and 9, the latter containing a pump 10, with a storage vessel 11 containing electrolyte and methanol or formic acid dissolved therein. With the help of the  
10 pump the electrolyte can be circulated in the closed circuit formed. By making the volume of the storage vessel large in relation to the volume of the electrolyte chamber, the fuel cell can be kept in operation for a long time  
15 without renewal of the electrolyte or supply of fresh fuel.

The electrolyte chamber 6 is connected to an evacuated container 12 by a conduit 13 containing an openable and closable valve 14.  
20 A gas chamber 16 above the liquid in the storage vessel 11 is in communication through a conduit 15 and the conduit 13 with a gas chamber 17 above the liquid in the electrolyte chamber 6. The pressure difference across  
25 the air electrode 3 can be read on a pressure gauge 18 which is connected to the conduit 13 by a conduit 19. The container 12 is provided with a conduit 20 at the bottom containing a valve 21 for removal of condensed products which may be formed.  
30

The container 12 can be evacuated, to a pressure corresponding to the vapor pressure above the electrolyte, by a pump 22 which is in communication with the conduit 13  
35 through a conduit 23 and an openable and closable valve 24. The pump 22 is powered by energy generated in the fuel cell, a driving motor 32 for the pump being connected to a storage battery 33 which is charged by the  
40 fuel cell. When the evacuation is completed, the valve 24 is closed and the valve 14 opened, if it has not already been opened during the latter part of the evacuation. The pressure in the container 11, the electrolyte  
45 chamber 6 and the container 12 is therefore equalised. If the container 12 has a large volume in relation to the gas chamber in the electrolyte chamber 6, the fuel cell may operate for a long time before reevacuation is  
50 necessary. Reevacuation of the container 12 may occur not only in connection with exchanging the electrolyte and fuel, but also during operation.

If the pressure gauge 18 is arranged in an electric control circuit in such a way that at certain pressures in the electrolyte chamber a signal is emitted to the motor 32 of the vacuum pump 22, and to the valve 24, so that the pump 22 is started and the valve 24  
60 is opened at a first pressure and the pump 22 stopped and the valve 24 closed when the required vacuum has been achieved, the device shown in Figure 1 can be made to operate automatically. The signal from the  
65 pressure gauge 18 is also arranged to in-

fluence the valve 14 so that this is closed at the first-mentioned pressure and reopened when the container has been evacuated.

In the device shown in Figure 2 (in which the same reference numerals used in Figure 1  
70 have been used to designate integers common to both Figures), a fuel electrode 25 consists of a porous layer 26 of the same material as is employed for the fuel electrode 2 and having the same pore structure as the layer 4  
75 in the device shown in Figure 1. On this layer is arranged a layer 27 with finer pores. This layer 27 is of the same type as the layer 5 on the air electrode shown in Figure 1. Outside the fuel electrode is a gas chamber  
80 28 which is supplied with a gaseous fuel at atmospheric pressure, for example hydrogen gas, from a container (not shown in the drawing), through an inlet 29. Unconsumed gas at the fuel electrode leaves through an outlet 30.  
85 An electrolyte chamber 31 is in its entirety situated between the fuel electrode 25 and the air electrode 3. Evacuation of the container 12 is carried out in the same way as has been described with reference to Figure  
90 1, as is also the achievement of the required difference pressure across the two electrodes.

The active layers 2 and 26 of the fuel electrodes, may be of other metals than nickel such as cobalt and iron activated with platinum metals, or for example, a Raney metal such as Raney cobalt or Raney nickel,  
95 in which case the electrode material usually contains a carrier material such as nickel or cobalt.

Instead of employing a mixture of nickel and silver for the active layer 4 of the air electrode, silver, platinum, palladium, platinum-plated carbon or manganese dioxide,  
100 may for example, be used.

The inactive layers 5 and 27 may, in place of the exemplified material, consist of a ceramic material such as aluminium oxide, aluminium silicate, zirconium oxide or thorium oxide.  
110

#### WHAT WE CLAIM IS:—

1. A fuel cell device comprising at least one fuel cell having a fluid electrolyte arranged in an electrolyte chamber, a fuel electrode arranged in contact with the electrolyte and  
115 with the fuel of the fuel cell and an air electrode arranged in contact with the electrolyte and with the atmosphere, the air electrode having one side contacting the atmosphere and one side contacting the electrolyte  
120 and comprising at least two porous layers having different porosity, the layer situated nearest the electrolyte having finer pores than the layer situated nearest the atmosphere, the electrolyte chamber being connected by an  
125 openable and closable conduit to an evacuated container which can be maintained at a pressure lower than atmospheric pressure by means of an evacuating pump connected to



- the container by an openable and closable conduit, the pump being arranged to be operated intermittently by electric energy generated in the fuel cell, the electrolyte being  
5 arranged in an electrolyte circuit containing an electrolyte storage vessel having a volume which is large in relation to that of the electrolyte chamber.
2. A fuel cell device according to claim 1,  
10 in which the fuel is dissolved in the electrolyte.
3. A fuel cell device according to claim 1 or 2, in which the fuel consists of methanol.
4. A fuel cell device according to claim 1  
15 or 2, in which the fuel consists of formic acid or a formate.
5. A fuel cell device according to claim 1, in which the fuel is gaseous, the fuel electrode  
having one side facing the fuel and one side facing the electrolyte and the fuel electrode 20 comprising at least two porous layers with different porosity, the layer situated nearest the electrolyte side having finer pores than the layer situated nearest the fuel side.
6. A fuel cell device substantially as here- 25 inbefore described with reference to, and as illustrated in, Figure 1 or Figure 2 of the accompanying drawing.

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COMPLETE SPECIFICATION

1 SHEET

This drawing is a reproduction of  
the Original on a reduced scale

Fig. 1

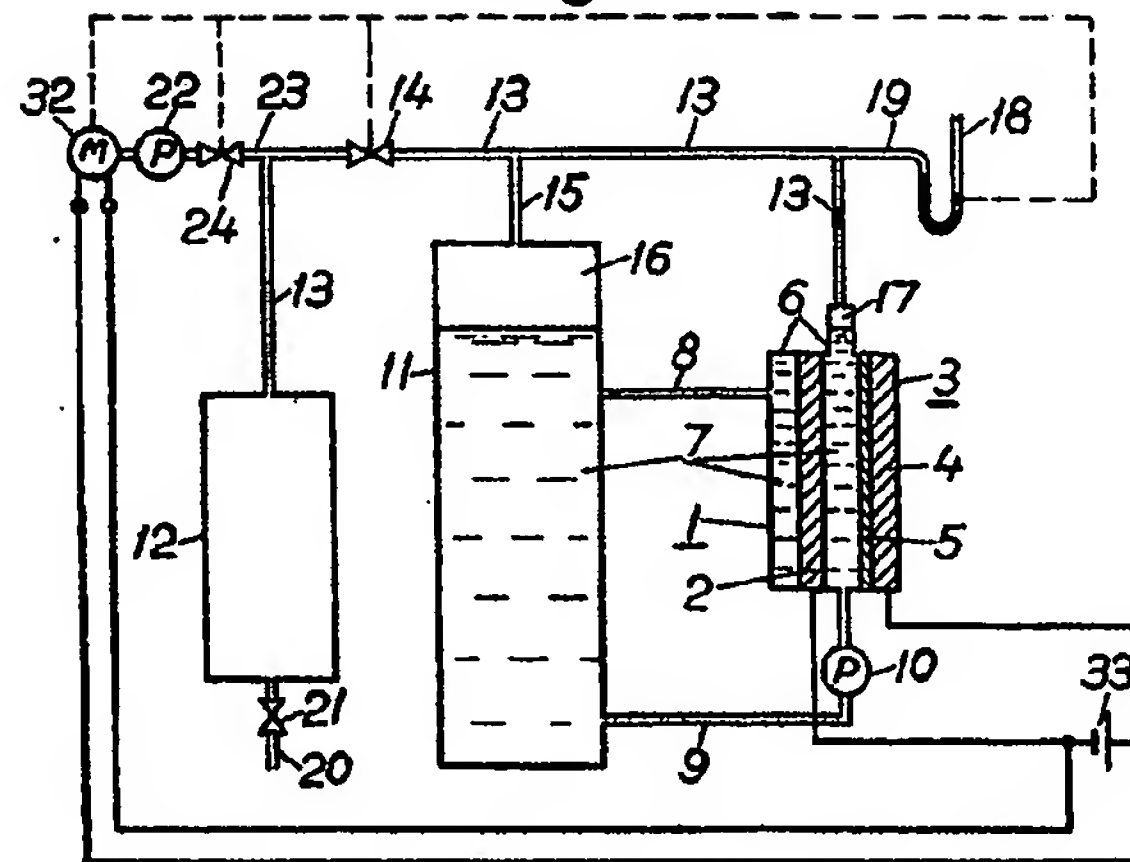


Fig. 2

